



PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE HONORABLE BOARD OF PATENT APPEALS AND INTERFERENCES

In re the Application of: Johannes Wilhelmus Maria Sonnemans, et al.

Application No.: 09/942,830

Filed: August 30, 2001

Docket No.: ACH 2823 US

Examiner: Tam M. Nguyen

For: PROCESS FOR EFFECTING ULTRA-DEEP HDS OF HYDROCARBON  
FEEDSTOCKS

BRIEF ON APPEAL

Appeal from Group 1764

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I. INTRODUCTION

This is an appeal from a final Office Action for the above-mentioned patent application, mailed June 28, 2005 rejecting claims 1, 7-12 and 18-28, which are all of the claims in the application.

A. Real Party in Interest

The real party in interest for this appeal and the present application is Albemarle Netherlands B.V. The assignment showing ownership by Albemarle Netherlands B.V. is recorded in the U.S. Patent and Trademark Office at Reel 016735, Frame 0113.

B. Statement of Related Appeals And Interferences

There are presently no appeals or interferences, known to Appellants, Appellants' representatives of the Assignee, which will directly affect or be directly affected by or have a bearing upon the Board's decision in the pending appeal.

C. Status of Claims

Claims 1, 7-12 and 18-28 are pending and are set forth in the attached Appendix. All of these claims stand finally rejected and are on appeal.

D. Status of Amendments

No amendment was filed after the final rejection forming the subject of this appeal.

E. Summary of Claimed Subject Matter

Independent claim 1 defines an ultra-deep hydrodesulfurization process for reducing the content of alkylated benzothiophenes and other sulfur compounds in a hydrocarbon feedstock

having certain characteristics including a sulfur content between about 150 ppm and about 500 ppm. The process reduces the sulfur content of the hydrocarbon feedstock to less than about 50 ppm. The process comprises contacting the hydrocarbon feedstock with a catalyst comprising: 1) a Group VIB metal component; 2) a Group VIII metal component selected from the group consisting of nickel, cobalt and iron; and 3) a sulfur-containing organic additive comprising a mercaptocarboxylic acid represented by the formula  $\text{HS-R1-COOR}$ , wherein R1 is a divalent hydrocarbon group with 1 to 10 carbon atoms and R is hydrogen, an alkali metal, an alkaline earth metal, ammonium, or a linear or branched alkyl group having 1 to 10 carbon atoms. Claim 1 also contains limitations regarding the hydrodesulfurization process conditions, namely, temperature, hydrogen partial pressure, liquid hourly space velocity and hydrogen/oil ratio.

Claims 7 through 11 depend upon claim 1 and further limit the hydrodesulfurization process conditions.

Independent claim 12 recites all of the limitations of claim 1 with the addition that the catalyst be subjected to a sulfidation and/or activation step before contacting the hydrocarbon feedstock.

Similar to claims 7-11, claims 13-22 depend upon claim 12 and further limit the hydrodesulfurization process conditions.

Independent claim 23 defines a two-step ultra-deep hydrodesulfurization process wherein a hydrocarbon feedstock having a sulfur content, including alkylated benzothiophenes, of about 0.1 wt% and not greater than 2 wt% is desulfurized into a product having a sulfur content of about 50 ppm or less. The process comprises a first step of contacting the hydrocarbon feedstock with a first catalyst, thereby reducing the sulfur content below about 500 ppm, followed by contact with a second catalyst, thereby reducing the sulfur content below about 50 ppm, wherein at least the second catalyst is characterized by the same limitations as recited in claim 1.

Claims 24 and 25 depend upon claim 23. Claim 24 adds the process limitation of fractionation or intermediate phase separation between contacting with the first and second

catalysts. Claim 25 adds the limitations that: 1) the first catalyst comprises molybdenum and cobalt and/or nickel; and 2) the second catalyst comprises molybdenum and nickel.

Independent Claim 26 recites all of the limitations of claim 23 with the addition that the first and/or second catalyst be subjected to a sulfidation and/or activation step before contacting the hydrocarbon feedstock.

Claims 27 and 28 depend upon claim 26. Claim 27 adds the process limitation of fractionation or intermediate phase separation between contacting with the first and second catalysts. Claim 28 adds the limitations that: 1) the first catalyst comprises molybdenum and cobalt and/or nickel; and 2) the second catalyst comprises molybdenum and nickel.

F. Grounds of Rejection to be Reviewed on Appeal

The two grounds for rejection are: 1) Claims 1, 7-12 and 18-28 are obvious over U.S. Patent No. 6,251,263 (Hatanaka et al.) in view of either U.S. Patent No. 4,845,068 (Takahashi et al.), EP 0357295 A2 (Takahashi), or EP 289211 A1 (Takahashi), collectively referred to herein as the "Takahashi references"; and 2) Claims 1, 7-12 and 18-28 are obvious over U.S. Patent No. 5,935,420 (Baird et al.) in view of either of the Takahashi references.

II. ARGUMENT

A. Rejection as Obvious over Hatanaka et al. in view of the Takahashi References

The crux of this appeal relates to the fundamental differences between the ultra-deep hydrodesulfurization (HDS) process as compared to conventional HDS. Generally speaking, HDS involves the removal of sulfur (as hydrogen sulfide) from various sulfur-containing compounds. In conventional HDS, the sulfur content of a hydrocarbon feedstock is reduced from a value of between 0.5 and 2 weight percent to a value of about 0.15 weight percent, which corresponds to 1500 ppm. The sulfur-containing compounds that react in a conventional HDS

process include sulfides, disulfides, thiophenes, and benzothiophenes. Sulfur is removed from these compounds with relative ease by a direct sulfur extraction reaction mechanism.

On the other hand, in ultra-deep HDS, the sulfur content of a hydrocarbon feedstock is reduced from a value of less than about 500 ppm to a value below about 50 ppm. The primary sulfur-containing compounds that must be reacted are alkylated benzothiophenes; and the reaction mechanism by which they react is believed to involve hydrogenation followed by sulfur extraction. The reacting compounds and reaction mechanisms for the two processes are so different that the catalysts one skilled in the art would expect to provide good results in a conventional HDS process do not perform as well in ultra-deep HDS. Similarly, catalysts that one skilled in the art may have believed to be inferior in a conventional HDS process exhibit surprisingly good results in ultra-deep HDS. It is for precisely this reason that the Examiner's proposed combination of references cannot establish a prima facie case of obviousness.

The Hatanaka et al. reference discloses a single-step hydrodesulfurization of a sulfur-containing feedstock in a reactor having three zones. Different hydrotreatment catalysts are present in each zone. The catalyst in the first zone consists essentially of cobalt and molybdenum supported on an alumina carrier. The catalyst in the second zone consists essentially of nickel and tungsten supported on an alumina carrier or zeolite-containing amorphous silica/alumina. The catalyst in the third zone consists essentially of cobalt and/or nickel, and molybdenum supported on an alumina carrier. The feedstock used in the Hatanaka reference has a sulfur content of, for example, 1.53 weight percent that is reduced to 240 ppm (Example 1); 230 ppm (Example 2); and 30 ppm (Example 3), respectively. Even though the feed sulfur content is greater than 500 ppm, the Hatanaka reference concerns ultra-deep HDS as the product sulfur is less than 50 ppm, at least in some of the examples. However, the reference does not disclose a catalyst comprising a sulfur-containing organic additive.

The Takahashi references all teach a similar hydrodesulfurization process, wherein the degree of desulfurization is reported as the "desulfurization rate." The highest desulfurization rate disclosed in the references is 93.5 percent, which means that 6.5 percent of the original sulfur content remained after processing the feedstock. *See* US 4,845,068, Col. 7, ln 35. In two

of the Takahashi references, the original sulfur content is 1.61 weight percent and the final sulfur content would be about 1000 ppm assuming the highest disclosed desulfurization rate. *See* US 4,845,068, Col. 5, ln 30, and EP 0289211, pg 5, ln 21. In the last Takahashi reference, the original sulfur content is 1.13 weight percent and the final sulfur content would be about 1500 ppm assuming the highest disclosed desulfurization rate. *See* EP 0357295, pg 9, ln 25. Given these sulfur ranges, it should be clear that the Takahashi references concern conventional HDS and not ultra-deep HDS.

The teaching, suggestion or motivation to combine the disclosures of prior art references is an essential evidentiary component of an obviousness holding. *C.R. Bard, Inc. v. M3 Sys., Inc.*, 157 F.3d 1340, 1352, 48 USPQ2d 1225, 1232 (Fed. Cir. 1998). Because the Hatanaka and Takahashi references concern fundamentally different processes, it simply cannot be concluded that one skilled in the art would be motivated to select and combine them.

Even if the teachings of these references could be combined, the combination would not lead to the process of the present invention. The Hatanaka reference utilizes three different catalysts, each used in a different reactor zone to address the problems encountered when desulfurizing a feedstock to below 50 ppm. *See* Hatanaka et al., Col. 4, ln 64 to Col. 5, ln 60. The Takahashi references only address the desulfurization of a feedstock having between 1 and 2 weight percent sulfur, which would correspond to the conditions present in the first reactor zone of the Hatanaka process. Thus, the combination of the two references, if it could be made, may lead one to replace the catalyst in the first reactor zone with the catalyst disclosed by Takahashi. There is no logical basis for replacing the catalysts in the other two reactor zones because Takahashi does not in any way suggest that this catalyst might be suitable for ultra-deep HDS. Furthermore, replacing the catalyst in the remaining reactor zones with a Takahashi catalyst would essentially reduce the Hatanaka reference to a single-zone process and would render the invention disclosed in Hatanaka inoperable for its intended purpose. *In re Gordon*, 733 F.2d 900, 902, 221 USPQ 1125, 1127 (Fed. Cir. 1984) (finding no suggestion to modify a prior art device where the modification would render the device inoperable for its intended purpose.) This resultant inoperable prior art reference should be considered to teach away from the proposed combination and should serve as further evidence of nonobviousness.

B. Rejection as Obvious over Baird et al. in view of the Takahashi References

As stated above, the Takahashi references do not concern ultra-deep HDS and are not properly combined with any reference concerning ultra-deep HDS, including the Baird reference. For this reason alone, the obviousness rejection cannot be maintained. However, even if the references could be combined, the combination would not lead to the process of the present invention.

The Baird reference recognizes that certain derivatives of dibenzothiophene are extremely difficult to remove from a hydrocarbon feedstock by conventional HDS processes and catalysts. The reference discloses a new process involving the use of a supported noble metal catalyst, wherein the noble metal is selected from the group consisting of platinum, palladium, iridium, and rhodium. The new process concerns reducing the sulfur content of a hydrocarbon feedstock to a product that has "substantially no sulfur," which is defined as less than about 1 ppm. See US 5,935,420, Col. 3, ln 34-42. In addition to noble metals, the catalyst system also comprises a hydrogen sulfide sorbent material. Thus, the reference teaches away from using cobalt/molybdenum or nickel/molybdenum catalysts. Like Hatanaka, Baird does not disclose a catalyst comprising a sulfur-containing organic additive.

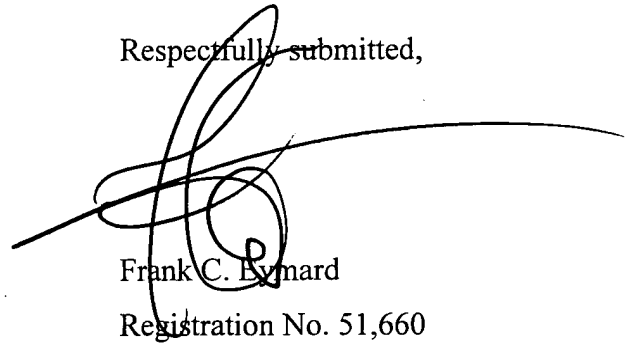
As with Hatanaka, even if the teachings of the Baird and Takahashi references could be combined, the combination would not lead to the process of the present invention. If the Baird process were used in conjunction with the Takahashi catalyst, the result would be inoperable. The Baird process employs a noble metal catalyst system together with a hydrogen sulfide absorbent to reduce sulfur content of a hydrocarbon feedstock to below 1 ppm. The Takahashi references do not suggest that one could achieve this level of sulfur reduction. Thus, this inoperable prior art reference should be considered to teach away from the proposed combination and should serve as further evidence of nonobviousness.



III. CONCLUSION

For all the reasons discussed above, it is respectfully submitted that claims 1, 7-12, and 18-28 define patentable subject matter under 35 U.S.C. § 103, and Appellants respectfully request this Honorable Board to reverse their rejection.

Respectfully submitted,

A large, stylized handwritten signature in black ink, appearing to read 'F. Byrard', is written over the typed name and registration number. The signature is fluid and cursive, with a long horizontal stroke extending to the right.

Frank C. Byrard  
Registration No. 51,660

APPENDIX

1. An ultra-deep hydrodesulfurization process for reducing the content of sulfur compounds comprising alkylated benzothiophenes in a hydrocarbon feedstock having an initial boiling point of not less than about 1000e and a 95% boiling point of about 4500e or less and a sulfur content between about 150 ppm and about 500 ppm to a sulfur content of less than about 50 ppm, comprising contacting said feedstock with a catalyst comprising a Group VIB metal component, a Group VIII metal component selected from the group consisting of nickel, cobalt and iron, and an S-containing organic additive comprising a mercaptocarboxylic acid represented by the general formula HS-R1-COOR, wherein R1 stands for a divalent hydrocarbon group with 1 to about 10 carbon atoms and R stands for a hydrogen atom, an alkali metal, an alkaline earth metal, ammonium, or a linear or branched alkyl group having 1 to about 10 carbon atoms, at a temperature from about 200 to about 450°C, a hydrogen partial pressure from about 5 to about 200 bar, a liquid hourly space velocity from about 0.1 to about 10 vol./vol.h and an H<sub>2</sub>/oil ratio from about 50 to about 2000 NI/l, thereby decomposing said sulfur compounds.

2. (canceled)

3. (canceled)

4. (canceled)

5. (canceled)

6. (canceled)

7. The process of claim 1, wherein said feedstock is contacted with said catalyst at a temperature from about 280 to about 430°C.

8. The process of claim 1, wherein said hydrogen partial pressure is from about 10 to about 100 bar.

9. The process of claim 1, wherein said hydrogen partial pressure is from about 15 to about 60 bar.
10. The process of claim 1, wherein said liquid hourly space velocity is from about 0.5 to about 4 vol./vol.h.
11. The process of claim 1, wherein said H<sub>2</sub>/oil ratio is from about 80 to about 1000 NI/l.
12. An ultra-deep hydrodesulfurization process for reducing the content of sulfur compounds comprising alkylated benzothiophenes in a hydrocarbon feedstock having an initial boiling point of not less than about 1000e and a 95% boiling point of about 450°C or less and a sulfur content between about 150 ppm and about 500 ppm to a sulfur content of less than about 50 ppm, comprising contacting said feedstock with a catalyst at a temperature from about 200 to about 450°C, a hydrogen partial pressure from about 5 to about 200 bar, a liquid hourly space velocity from about 0.1 to about 10 vol./vol.h and an H<sub>2</sub>/oil ratio from about 50 to about 2000 NI/l, said catalyst comprising a Group VIB metal component, a Group VIII metal component selected from the group consisting of nickel, cobalt and iron, and an S-containing organic additive comprising a mercaptocarboxylic acid represented by the general formula HS-R1-COOR, wherein R1 stands for a divalent hydrocarbon group with 1 to about 10 carbon atoms and R stands for a hydrogen atom, an alkali metal, an alkaline earth metal, ammonium, or a linear or branched alkyl group having 1 to about 10 carbon atoms, said catalyst being subjected to a sulfidation step and/or activation step before contact with said feedstock, thereby decomposing said sulfur compounds.
13. (canceled)
14. (canceled)
15. (canceled)
16. (canceled)

17. (canceled)

18. The process of claim 12, wherein said feedstock is contacted with said catalyst at a temperature from about 280 to about 430°C.

19. The process of claim 12, wherein said hydrogen partial pressure is from about 10 to about 100 bar.

20. The process of claim 12, wherein said hydrogen partial pressure is from about 15 to about 60 bar.

21. The process of claim 12, wherein said liquid hourly space velocity is from about 0.5 to about 4 vol./vol.h.

22. The process of claim 12, wherein said H<sub>2</sub>/oil ratio is from about 80 to about 1000 NI/l.

23. A two-step ultra-deep desulfurization process for converting a starting feedstock having an initial boiling point of not less than about 1000G and a 95% boiling point of about 450°C or less and having a sulfur content comprising alkylated benzothiophenes of above about 0.1 wt. % and not greater than about 2 wt. % into a product having a sulfur content of about 50 ppm or less, wherein the process comprises contacting said feedstock with a first catalyst followed by contact with a second catalyst, both catalysts comprising a Group VIB metal component and a Group VIII metal component selected from the group consisting of nickel, cobalt and iron, with at least said second catalyst additionally comprising an S-containing organic additive comprising a mercaptocarboxylic acid represented by the general formula HS-R<sub>1</sub>-COOR, wherein R<sub>1</sub> stands for a divalent hydrocarbon group with 1 to about 10 carbon atoms and R stands for a hydrogen atom, an alkali metal, an alkaline earth metal, ammonium, or a linear or branched alkyl group having 1 to about 10 carbon atoms, the conditions for said contact with both catalysts being the same or different and comprising a temperature from about 200 to about 450°C, a hydrogen partial pressure from about 5 to about 200 bar, a liquid hourly space velocity from about 0.1 to

about 10 vol./vol.h and an H<sub>2</sub>/oil ratio from about 50 to about 2000 NI/l, the effluent from contact with said first catalyst having a sulfur content of less than about 500 ppm, and the product after contact with the second catalyst having a sulfur content of less than about 50 ppm.

24. The process of claim 23, wherein the effluent following contact with said first catalyst is contacted with said second catalyst after fractionation or intermediate phase separation.

25. The process of claim 23 wherein the first catalyst comprises molybdenum as Group VIB metal component and cobalt and/or nickel as Group VIII metal component, while the second catalyst comprises molybdenum as Group VIB metal component and nickel as Group VIII metal component.

26. A two-step ultra-deep hydrodesulfurization process for converting a slalling-feedstock having an initial boiling point of not less than about 100°C and a 95% boiling point of about 450°C or less and having a sulfur content comprising alkylated benzothiophenes of above about 0.1 wt. % and not greater than about 2 wt. % into a product having a sulfur content of about 50 ppm or less, wherein the process comprises contacting said feedstock with a first catalyst followed by contact with a second catalyst, the conditions for said contact with both catalysts being the same or different and comprising a temperature from about 200 to about 450°C, a hydrogen partial pressure from about 5 to about 200 bar, a liquid hourly space velocity from about 0.1 to about 10 vol./vol.h and an H<sub>2</sub>/oil ratio from about 50 to about 2000 NI/l, the effluent from contact with said first catalyst having a sulfur content of less than about 500 ppm, and the product after contact with the second catalyst having a sulfur content of less than about 50 ppm, both of said catalysts comprising a Group VIB metal component and a Group VIII metal component selected from the group consisting of nickel, cobalt and iron, with at least said second catalyst additionally comprising an S-containing organic additive comprising a mercaptocarboxylic acid represented by the general formula HS-R1-COOR, wherein R1 stands for a divalent hydrocarbon group with 1 to about 10 carbon atoms and R stands for a hydrogen atom, an alkali metal, an alkaline earth metal, ammonium, or a linear or branched alkyl group having 1 to about 10 carbon atoms, said first catalyst and/or said second catalyst being subjected

to a sulfidation step and/or activation step before contact, respectively, with said feedstock or contact with the effluent from contact with said first catalyst.

27. The process of claim 26, wherein the effluent following contact with said first catalyst is contacted with said second catalyst after fractionation or intermediate phase separation.

28. The process of claim 26 wherein the first catalyst comprises molybdenum as Group VIB metal component and cobalt and/or nickel as Group VIII metal component, while the second catalyst comprises molybdenum as Group VIB metal component and nickel as Group VIII metal component.